Energy 45 (2012) 817–828

Contents lists available at SciVerse ScienceDirect

Energy

journal homepage: www.elsevier.com/locate/energy

System aspects of biomass gasification with methanol synthesis – Process concepts and energy analysis

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ARTICLE INFO

Article history:
Received 27 February 2012
Received in revised form
29 June 2012
Accepted 3 July 2012
Available online 31 July 2012

Keywords:
Biomass gasification
Methanol synthesis
Energy balance
Process integration

ABSTRACT

A systematic analysis of interrelations between different process steps in the biomass gasification and methanol synthesis chain was made. The energy performance of the system is assessed by reviewing previous studies and analysing a case study. Implications of technology choices and process integration opportunities are in focus.

The case study analyses the impact on the energy balance of process-integrated drying compared to import of dried biomass, effects on electricity production due to heat pump integration or district heating (DH) delivery, effect on methanol yield of hydrogen addition and the impacts of adding an methanol-to-olefins (MTO) process.

The biomass drying has significant impact on the overall energy balance, the cooling demand increases by 60% for the case study installation when process-integrated drying is not applied for drying the biomass. There is a trade-off between methanol yield, electricity and DH production potential of the system. Heat pumping can increase the electricity yield of the system. Hydrogen addition replacing the water gas shift can increase the methanol yield, in our case study by ~35%, but in a stand-alone case the electricity demand makes such a system unrealistic. Adding an MTO unit to the system has limited impact on the energy balance.

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1. Introduction

Biomass gasification is seen as one of the key technologies for the replacement of fossil fuels in the transport sector, and the application of the technology has significant potential for co-production of several products such as fuels, chemicals, materials, electricity and heat. The biomass gasification technology is still not commercialised in full scale but several demonstration plants are under way, and hence studies aimed at developing energy-efficient, economically viable and resource-efficient systems are necessary. Many previous studies on this subject have been devoted to optimising specific process steps in the gasification or synthesis processes; some have included the entire conversion system and integration possibilities, but few studies have analysed the entire system and its interactions with incumbent industry and infrastructure [1].

In this paper the energy balance for a commercial-scale biomass gasification system producing methanol or materials is analysed. Different process integration possibilities and uses for excess heat are investigated. The results will be used in future studies investigation other types of integration.

2. Objective

In order to evaluate the importance of integration possibilities with other industries and infrastructure, good knowledge of a comparative stand-alone system is necessary. The aim of this paper is to identify a technical solution for a biomass gasification system with methanol synthesis and investigate the stand-alone energy balance of such a system. In addition to technology choices of process units, the impact on the energy balance of process-integrated drying compared to import of dried biomass,

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Abbreviations: ASU, air separation unit; ATR, autothermal reformer; CC, combined cycle; CFB, circulating fluidized bed; DH, district heating; DME, dimethyl ether; EF, entrained flow; FICFB, fast internal circulating fluidized bed; GCC, grand composite curve; GT, gas turbine; HGC, hot gas cleaning; LPMeOH, liquid phase methanol synthesis; MEA, monoethanolamine; MeOH, methanol; MTO, methanol-to-olefins; PSA, pressure swing absorption; SMR, steam methane reformer; SNG, synthetic natural gas; ST, steam turbine; WGS, water–gas-shift.

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0360-5442/$ – see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.energy.2012.07.009

and effects on electricity production due to heat pump integration or district heating (DH) delivery, are investigated. This paper includes two concepts in addition to the conventional systems: one where the methanol synthesis is followed by an methanol-to-olefins (MTO) process which produces ethylene and propylene, and another where an electrolyser produces hydrogen and oxygen replacing the water gas shift and some of the air separation unit (ASU) capacity.

This paper also includes a systematic analysis of interrelations between process steps in the gasification and synthesis chain, and implications of different technology choices in terms of process integration opportunities and energy efficiency.

The studied woody biomass gasification system in the case study was identified based on the criteria that it should be of reasonable commercial-scale (e.g. ~400 MWth input) and technically feasible to implement within 5–10 years.

3. Methods

A review of studies evaluating biomass gasification systems with downstream methanol synthesis was made and a systematic analysis of the results of the studies was performed. Only studies with detailed information on technology choices for the different process steps were included. Based on the review, a conversion route from biomass to methanol, which could serve as an example in order to evaluate the stand-alone energy balance and the impacts of important integration and boundary conditions, was identified. Some important process integration issues have already been described in the reviewed studies, but some additional aspects – such as whether the biomass is dried on- or off-site; trade-off between fuel, electricity and hot water/steam production; and additional further upgrading – are illustrated by examples in this study.

The systematic analysis of the interrelationships of the different process steps in the gasification and synthesis chain includes upstream processes, e.g. biomass pre-treatment, and downstream process, e.g. synthesis reaction and final products. The analysis includes implications of technology choices and process integration opportunities. Many of the conclusions from the analysis apply for biomass gasification systems with other downstream synthesis reactions, like Fischer–Tropsch, synthetic natural gas (SNG) and dimethyl ether (DME). Literature describing studies of such systems was also consulted, but focus and references in this paper are limited to key references; reviews [1–3] and others [4–6] and references on methanol production systems based on gasification of lignocellulosic biomass.

The energy performance and process integration possibilities of the methanol production systems are analysed using pinch technology. Pinch technology is a combination of pinch analysis and process design based on pinch rules. This technology is widely used for determining minimum heating and cooling demands of industrial processes and identifying potential improvements and integration possibilities. Pinch analysis was originally developed by Linnhoff [7]. Previous studies of process industry have shown that this is a strong tool for identifying process integration possibilities [8–10].

Information on heat balances of the different process streams in the biomass conversion system used as example in this study is based on modelling performed by Isaksson [11].

4. System aspects of biomass gasification configurations

A biomass gasification system is a system with many interrelationships both within the industrial installation and with the surrounding society and environment. Table 1 lists the most important technical (and physical) factors and their interdependences. The different choices made for each of the factors result in different efficiency (energy and environmental) and economy.

The gasification process results in significant amounts of excess heat, and the ability of the system to make use of this excess heat (at different temperature levels) will be of great importance for the overall energy balance and economic and environmental performance of the system. This study is limited to the energy balance of stand-alone systems, although some general comments on the economic performance are given.

In addition to the already mentioned factors, there are always non-technical and non-economic factors that will have an impact and that need to be considered for large-scale industrial settings: e.g. social (business relations, available manpower and know-how, acceptance, etc.), legal (emission limits etc.), practical (timing, uptimes etc.) and safety. However, those are not analysed in this study.

4.1. Components in the gasification system

Different biomass gasification technologies are differently suited for various sizes, raw materials and downstream synthesis reactions. The requirement for pre-treatment of the biomass also differs. Therefore, consideration of the reciprocal dependence between the process steps in the gasification chain needs to be taken when setting up a gasification system (see Fig. 1).

The most common classification factors for gasifiers are [2]: gasification agent; pressure; temperature; fluid dynamics and heat supply to the gasifier. The interdependence of the different classification factors on upstream and downstream units are further explained below.

4.1.1. Gasification agent

The choice of gasifying agent depends on, for instance, the gasification technology, the downstream utilisation of the product gas, and economy. The most common gasification agents are air, steam and oxygen (or mixtures thereof). Air is cheap and abundant but it contains significant amounts of N_2 and the product gas will be diluted. The syngas from air gasification will have a comparably low heating value (4–7 MJ Nm^{-3}). Many downstream applications require N-free gas and would thereby require costly N_2-separation. Steam is also abundant at most industrial sites although the production requires energy. Steam is often used as gasification agent together with oxygen. Pure oxygen could be produced on site by a conventional air separation unit, which is a highly energy-(electricity-) demanding process [3,4]. The product gas from oxygen or steam gasification will have a higher heating value (10–14 MJ Nm^{-3}). Using steam results in higher percentages of hydrogen, which increases the heating value of the product gas. However, using steam requires higher operating temperatures and thereby makes the process more expensive. A common solution is to use a mixture of air and steam.

4.1.2. Pressure

The main advantages of pressurising the process are that the equipment can be made smaller and, if the downstream process requires elevated pressures, compression energy could be saved. The CH_4 and CO_2 concentrations in the product gas increase with higher pressures in the gasifier, whereas the syngas concentration (H_2 and CO) is reduced. Higher pressure in the gasifier also means higher consumption of oxidising agent, and there is an upper limit for pressurisation at 70–100 bar for practical reasons of the equipment [4].

4.1.3. Temperature

In order to avoid agglomeration and to maintain good conditions for ash removal, there is, for each gasification technology,

Table 1
Interrelationships between elements in the biomass gasification system.

System element	Interrelated Parameter	Consequences/implications
Raw material	Raw material properties	Pre-treatment requirements (drying, comminution, etc.), syngas composition, location of pre-treatment (on- or off-site) can impact costs and energy demand for transportation. Economic distance for raw material supply can increase with some pre-treatments.
	Gasification technology	Pre-treatment requirements (e.g. torrefaction and pyrolysis). Pre-treatment processes result in energy losses.
	Size of gasifier	Supply possibilities in terms of availability and infrastructure
Localisation	Infrastructure	Raw material supply and end product distribution (harbour, railway, roads, natural gas net, DH net, etc.)
	Integration opportunities	Heat sinks/sources; DH net (low-temperature heat, high temporal variation in demand). Industry (other temperature levels, less time, variable demand). Other than heat-based integration opportunities.
	Scale	Amount of raw material in nearby area; transportation distances. Integration with incumbent industry and DH net. Technology choices.
Gasifier	Product gas composition	Downstream equipment (tar cleaning, reformer etc.)
	Downstream application	Gas cleaning requirements; optimal temperature and pressure level
	Gasifying agent	Oxygen produced in ASU, but expensive and energy-demanding. Air is cheap but results in N_2 dilution. Steam is cheap but adds much moisture. Only suitable for some technologies.
	Pressure	Pressurisation is advantageous for size of equipment and investment costs. Pressurisation complicates feeding of solid biomass and is difficult in the case of indirect gasification.
	Temperature	High temperature results in low CH_4 concentration in syngas. High temperatures reduce tar formation. Low temperatures favour high methane concentration and result in high tar concentrations.
	Raw material	Moisture content, particle size and chemical composition.
Gas cleaning/ conditioning	Downstream application	Specifications on contaminants and tars
	Gasification technology	Catalytic bed materials can reduce tars in low-temperature gasification technologies. High-temperature technologies result in low tars which imply no need for reforming.
Synthesis	Gasification technology	In the case of pressurised synthesis reactors it will be more efficient to also have pressurised gasification.
	Gas cleaning	Synthesis specifications determine which gas cleaning is needed.
	End products	Synthesis setup impacts the end product composition. Many synthesis reactions require CO_2-separation, hence CO_2 is a “product”.
Conditioning/ upgrading	End use	The end use of the products determines the level of conditioning and upgrading needed.
	Infrastructure	In the case of distributing DH or SNG by incumbent infrastructure (networks) these systems will set the requirements for upgrading.
End products	Technical conversion system[a]	The configuration of the gasification system and synthesis including conditioning and upgrading result in conversion efficiency for the end products: biofuels, chemicals, electricity, district heating.
	Incumbent industry and infrastructure	Possibilities to deliver/retrieve steam to/from industrial processes. Delivery and transport of e.g. DH or other products (SNG).

[a] Technical conversion system includes gasification technology, gas cleaning, synthesis and upgrading.

a raw material-specific restricted temperature interval between the softening and slagging temperatures of the ash [5]. The temperature also has a significant impact on the composition of the product gas: in most cases the product gas contains CO, H_2, CH_4, aliphatic hydrocarbons, benzene, toluene and tars as well as CO_2 and H_2O. In high-temperature gasification ($T > 1200$ °C), the biomass is completely converted into H_2, CO, CO_2 and H_2O and the gas is chemically similar to syngas derived from fossil sources.

4.1.4. *Fluid dynamics*

The fluid dynamics are related to the shape of the gasifier and how the biomass and the oxidising agent meet. The fluid dynamics set some restrictions on size of gasifier and biomass particles. Mainly three classes of gasifiers exist: fixed bed, fluidised bed and entrained flow.

4.1.5. *Heat supply to the gasifier*

Gasifiers can be heated both directly and indirectly. One advantage of indirectly heated gasifiers is that the gasifying agent will not come in contact with the product gas and hence air can be used without N_2 dilution. The disadvantage is the complexity of pressurisation and up-scaling.

4.2. *Gas cleaning and conditioning*

The gas cleaning requirements of the product gas depend on the gasification technology and the downstream applications. If the gasifier is low-temperature (<1000 °C) and the downstream application is not favoured by high methane concentrations, it is common to have a reformer in order to reduce the amount of methane in the product gas. Most synthesis reactions are sensitive to tars and impurities like sulphur. For some synthesis reactions it is also necessary to remove CO_2 (which normally is done by Selexol, Rectisol, MEA or PSA).

For tar removal there are several options: catalytic bed material in the gasifier and scrubbing are commonly used, but also catalytic reforming and chemical looping reforming are technologies under development [12].

4.3. *Synthesis application*

As described in the previous sections on gasification technology, the synthesis section is strongly linked to upstream conditions. Most synthesis reactions operate at specific temperatures and pressures and have high requirements on gas composition. Most

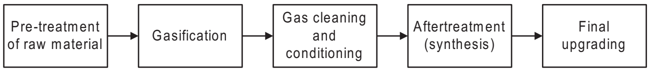
Pre-treatment of raw material
Gasification
Gas cleaning and conditioning
Aftertreatment (synthesis)
Final upgrading

Fig. 1. Schematic description of process steps in the biomass gasification chain.

synthesis catalysts are sensitive to tars, particulates, sulphur, CO_2 and other impurities. Commonly the gas passes a water–gas-shift (WGS) reactor in order to achieve the preferable H_2/CO ratio. If the gasification is operated at lower pressure levels than the required synthesis pressure, compression is needed. Upstream pressurisation will save energy.

Also the temperature level of preceding steps (e.g. gas cleaning) will impact the energy demand for the synthesis. Most syngas syntheses (methanol, Fischer–Tropsch, methanation etc.) are exothermic reactions, but the temperature level differs and also the recycle rate which impacts the possibilities for process integration and heat recovery. Synthesis reactions where CO_2 needs to be removed will add CO_2 to the co-products.

4.4. End products

It is the final syngas application that determines the specifications for the product gas, and hence also determines most of the setup of the gasification system.

If the syngas is used in an integrated gas combined cycle (IGCC) for electricity and heat production, the purification need is less stringent than for synthesis processes; however, the heating value needs to meet certain specifications.

If the syngas is used for synthesis of 2nd-generation biofuels or chemicals, more extensive cleaning and conditioning are required; see Section 4.3. This has an impact on the energy performance of the system and suitable choices for gasification technology and gas cleaning.

The gasification process results in significant amounts of excess heat, and the ability of the system to make use of this excess heat will be of great importance for the overall economy of the system. The heat can be used in different ways, such as:

- For the production of electricity (requires gas turbine, steam turbine or other electricity generation equipment).
- For process integration, for instance with downstream synthesis reactions, gas cleaning steps or biomass drying and pre-treatment.
- For production of district heating. This needs to be matched with a demand for district heating and a network connected to the gasification plant.
- For integration with other, co-located, industry.

4.5. Raw material

The efficiency in the gasification process is improved with the dryness of the raw material [6]. At the same time the hydrogen content of the syngas is reduced, which can be a disadvantage for certain downstream processes (e.g. Fischer–Tropsch and methanol synthesis). Since there is a cost for drying the biomass (each incremental drying adds to that cost) there will always be an economically optimal level of drying.

Since biomass is a bulky material and expensive to transport, decentralised pre-treatment such as drying, pyrolysis or torrefaction is sometimes suggested to reduce transportation cost. Torrefaction and pyrolysis also have positive effects on the cost for size reduction of the biomass. Pyrolysis liquefies the biomass which thereby is easy to pressurise and feed into the gasifier. Torrefaction results in solid biomass with very low moisture content and the material becomes brittle, and the cost for grinding is significantly reduced. The negative effect of pyrolysis and torrefaction is that there are losses involved, approximately 30% of energy in the case of pyrolysis and 10% in the case of torrefaction [13]. One possibility to efficiently supply the energy needed for these biomass pretreatments is to use excess heat from the gasification and synthesis reactions.

The size of the gasifier and the raw material supply can impact the pre-treatment choices. If the raw material can be supplied from the nearby area, pre-treatment might be done most efficiently at the gasification site. However, if biomass needs to be imported to the region, upgraded biomass might be a more realistic alternative. This will impact the price of the raw material, of course, and several studies show that the cost of raw material is an important part of the costs of a biomass gasification plant (e.g. [14]).

4.6. Localisation

4.6.1. Infrastructure for raw material supply and product distribution

Nohlgren et al. [15] argue that the raw material supply of a biomass gasification system should be available within a distance of 100–150 km; however, coastal localisations could increase this distance. The relation between distance, cost and transportation modes for biomass supply to biomass gasification systems was studied by Ekbom et al. [16]. Ship and truck transports are most efficient for long and short transport distances respectively.

For the utilisation of excess heat, the proximity to a district heating system or other heat sinks is of substantial importance. Leduc et al. [17] conclude that the DH demand and the heat price strongly affect the methanol production cost in a biomass to methanol plant located in northern Sweden. The product demand and the product distribution are also important to consider [17–19]. For the distribution of other products (other than liquid fuels and DH), for instance, bio-SNG a location in close connection to the natural gas grid is an important advantage, compared to a site with no net distribution possibility.

4.6.2. Integration opportunities

Integration possibilities within the gasification and synthesis system, as well as with other adjacent process industries, are important for the overall energy efficiency of the system. Previous studies of the energy performance of 2nd-generation fuel production (e.g. Fischer–Tropsch, DME, SNG, methanol etc.) via biomass gasification have in many cases been limited to part of the process chain, although there are studies considering the entire industrial process. Some of these studies have applied pinch methodology for improving and optimising the energy efficiency of systems with different technology solutions (e.g. [12,14,20–22]). There are also some studies that include the integration of the gasification process with other (incumbent) industrial systems/processes [8,11,23,24].

The examples given in this paper on biomass gasification systems with methanol production focus on stand-alone cases, but configurations and system boundaries are altered. This means that internal process integration is considered. Integration possibilities with incumbent industries are also important but such analysis is not included in this study.

4.6.3. Scale

In general, there are significant advantages of scale for biomass gasification units due to equipment costs. Concerning the cost for raw material supply, Boerrigter and van der Drift [25] show that the increased cost for transportation is less than the economic benefits of increased scale. Hence, the raw material cost is not a reason for keeping the unit smaller.

5. The biomass gasification system with downstream methanol synthesis

Biomass gasification systems with downstream methanol synthesis previously described in literature and analysed from energy efficiency perspective were reviewed. Some interrelationships and system boundary conditions not taken into consideration in previous studies were identified and are illustrated and analysed in the stand-alone case study in Section 5.3.

5.1. *Process description – biomass gasification with downstream MeOH synthesis*

In Fig. 2 a schematic process layout for the methanol production processes via biomass gasification is given. The figure indicates different alternatives for a number of process units in terms of optional steps and technology alternatives. Several recycle options and several options for heat recovery (including possibilities to use steam turbine for electricity generation) are indicated. The different options for gasification technologies are not indicated, but as discussed in Section 4.1 there are several technology options. The process steps marked as optional are not necessary to include in all configurations. For instance, drying is not needed if the raw material is dry enough (e.g. it has been dried elsewhere). Pyrolysis or torrefaction might be necessary for some gasification technologies (e.g. entrained flow) but not for all. Oxygen production is needed to supply oxygen and steam/oxygen-blown gasifiers. Reforming might be needed if the gasifier operates at lower temperatures and/or the downstream synthesis is sensitive to methane or heavier hydrocarbons. Many synthesis reactions are sensitive to CO_2 which makes removal necessary.

5.1.1. *Methanol synthesis*

Conventional methanol reactors are fixed bed reactors of catalyst pellets and operate in gas phase. There are different types but the subsequent heating and cooling leads to an inherent inefficiency [26]. The synthesis is sensitive to sulphur and CO_2 and these compounds have to be removed from the product gas before entering the synthesis step. The H_2/CO ratio should be close to 2 in order to maximise efficiency and yield. Methanol synthesis reactors operate within 50–150 bar pressure and 230–270 °C temperature. Levels around 250 °C and 100 bar are common and suitable [20].

In the 1980s, Air Technology developed a liquid phase methanol (LPMeOH) slurry reactor which has been demonstrated but is not used on a commercial-scale today [27]. The technology is still under development and shows good opportunities for increased efficiency. The process is insensitive to CO_2 and thereby no CO_2 removal is required before synthesis [26]. Operational conditions for the LPMeOH are 230–260 °C and 50–100 bar [28].

5.2. *Energy performance of biomass gasification systems with methanol synthesis*

In Fig. 3 examples of the energy efficiencies of different biomass to methanol systems presented in previous studies are shown. The efficiencies are not directly comparable due to differences in assumptions and delimitations in the studies. Hence Fig. 3 does not identify the most efficient system or even the system with the highest yield of methanol, but shows the variation in performance and outputs of different configurations. Details on the configurations and assumptions for the different systems are given in Appendix, and some of the general differences in assumptions and delimitations are explained below:

- The properties of the input biomass differ; e.g. the water content of the fuel is in most cases 50% but in some cases lower (see Appendix). Most studies consider biomass drying at the gasification plant, but the drying level differs. Moisture content at input to gasifier of 15–20% is common but more severe pretreatment is considered, such as pyrolysis or torrefaction is considered in some cases. The input biomass is solid in all cases except in scenarios 16–21, [20], which start with bio-oil (pyrolysis oil from poplar wood, with water content of 30% as received at the methanol plant).
- The assumptions concerning the power generating equipment differ; in some studies both a gas turbine (for off-gases) and a steam turbine (ST) are considered, whereas some studies only consider a steam turbine. Tock et al. [14] considers expansion turbines for off-gases to reach the same pressure as for the burner.
- The indicated "yield" of heat is not directly comparable since the quality (pressure and temperature) of the heat is not the same for all cases. In general the heat quality is at levels suitable for district heating, but in scenario 4 it is steam at 3.5 bar which is higher than required for DH.
- There are differences in purity of the end product (95–99% wt).

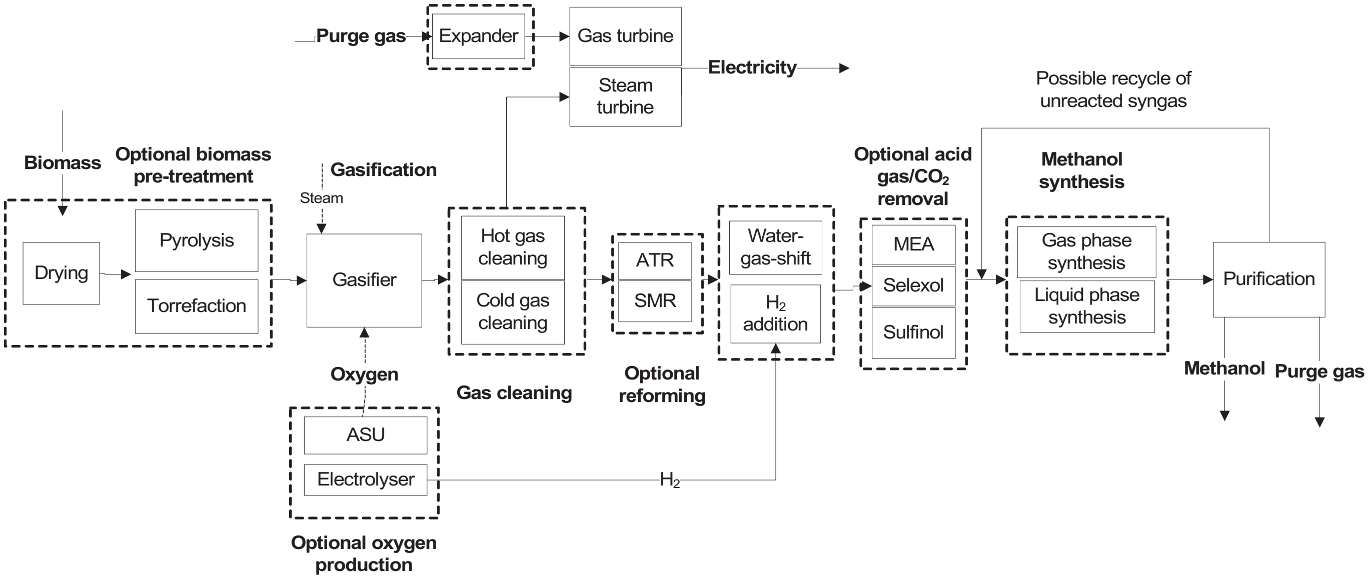

Fig. 2. Schematic process layout for biomass to methanol process indicating different system solutions.

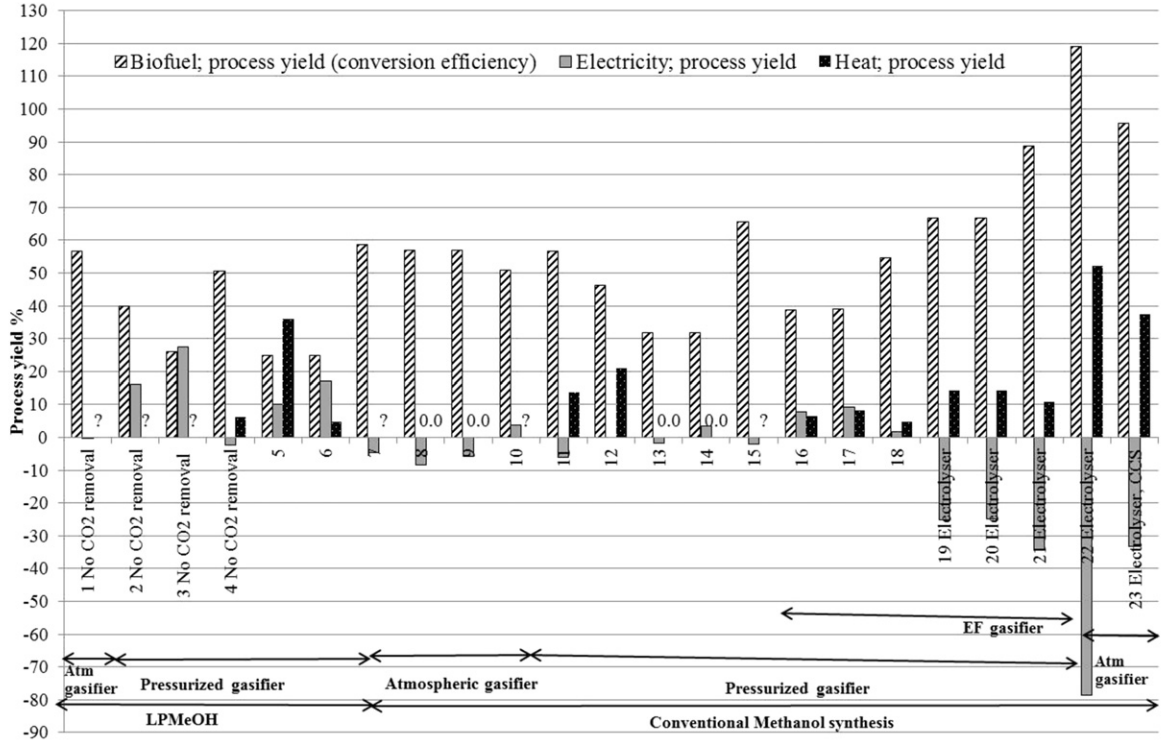

Fig. 3. Yields of methanol, electricity and heat (DH grade >100 °C) of biomass gasification systems with methanol synthesis from a number of studies, (see Appendix 1). The yields of the different studies are not comparable to each other due to differences in delimitations and system boundaries. The figure shows the range of results available in literature and the trade-off between methanol, electricity and heat production. Yields are based on LHV of input biomass.

Hamelinck et al. [26] (scenarios 1–3, 7 and 10 in Fig. 3) concluded from their comparison that, due to the lower energy quality of fuel compared to electricity, once-through systems perform better than systems aimed mainly for fuel production. Further, they say that hot gas cleaning can promote better performance. Their scenario with indirect gasification, wet gas cleaning, LPMeOH and recycle turned out to have the best economic performance.

Tock et al. [14] considered conventional methanol synthesis but different gasification technologies, indirect fast internal circulating fluidised bed (FICFB) and direct circulating fluidised bed (CFB) pressurised oxygen-blown, and different technologies for CO_2 removal, MEA and Selexol respectively (scenarios 8–9 and 13–14). According to their detailed process integration analysis, most of the methanol systems were net electricity consumers. They show that CO_2 removal by chemical absorption (MEA) is a large heat sink strongly influencing the energy integration, the combined heat and power generation, and hence the overall energy efficiency.

Isaksson et al. [11] (scenario 4) have investigated the possibility of integrating the production of methanol (via biomass gasification) with a pulp and paper mill. In addition to methanol and electricity production, the system delivers steam at 3.5 bar to the paper mill.

Brandberg et al. [29] (scenarios 5–6) considered the LPMeOH synthesis in a once-through mode, and the resulting methanol yield is very low compared to other studies. All the unconverted fuel gas was assumed to be used in the combined cycle for electricity production.

All scenarios in Fig. 3 with pressurised gasification are oxygen-blown or oxygen/steam-blown, and hence require oxygen production. All scenarios with atmospheric gasification are indirectly heated, except for scenarios 22–23, which are based on a directly heated two-stage technology. Most of the pressurised cases include CFB gasifiers, whereas [20] (scenarios 16–21) consider entrained flow gasification. Two studies [14,26] (scenarios 1, 7–9) include atmospheric gasification and the methanol yield is very similar in all four scenarios (57–59%), and the electricity deficit ranges from 0 to 8.5%. However, for the pressurised scenarios there is a greater variation in methanol yield ranging from 26 to 89%, which is mainly due to differences in recycle conditions. It can also be seen that scenarios with higher methanol yield generally have lower net electricity production.

The methanol yield of scenarios 19–23 [20,30] stands out due to the application of an electrolyser instead of an ASU. The electrolyser splits H_2O into O_2 and H_2 and the oxygen is used in the gasification process whereas the hydrogen is added to the syngas, improving the yield of methanol significantly. The drawback is the significantly increased electricity deficit of the system. Note that the yields presented in Fig. 3 are based on input biomass (LHV) and hence, do not include added hydrogen, which explains why the yield can be higher than 100%. Also other studies of bio-syngas to methanol systems conclude that hydrogen addition can improve the carbon conversion significantly [31]. Clausen et al. [30] uses a two-stage gasifier at atmospheric pressure which has a lower oxygen demand than the pressurised used by Sadhukhan and Ng [20]. In scenario 22 more oxygen than needed in the gasification process is produced by the electrolyser whereas in scenario 23 some of the CO_2 in the syngas is removed in order to not have any oxygen surplus (hence the H_2 addition is smaller).

Scenario 15 [32] does not include the H_2 addition but still has an exceptionally high methanol yield. In this setup a pressurised fluidised-bed gasifier is followed by a novel catalytic gas reforming. A high-temperature shift is applied which makes it possible to do the shift reaction before scrubbing, leading to smaller losses. In addition there is some recirculation from the shift reactor to the gasifier. All in all, this results in the high methanol yield and the comparatively low net electricity demand.

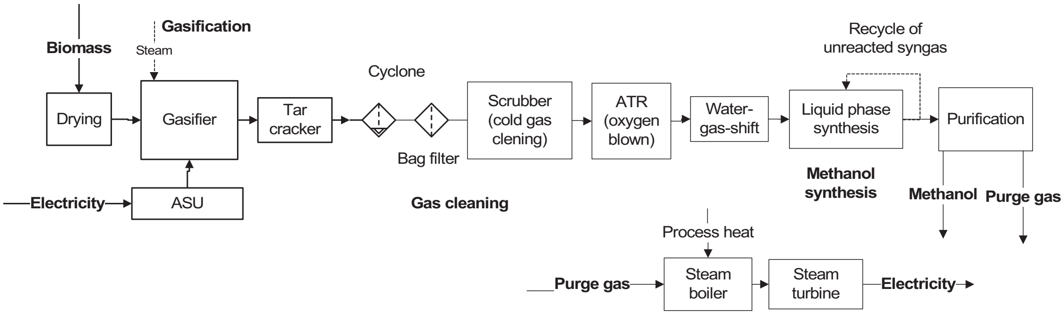

Fig. 4. Process description of stand-alone case study.

Since the methanol synthesis takes place at high pressures, 80–100 bar, systems with pressurised gasification will save compression electricity. Even though more electricity will be needed in the gasification step due to pressurisation, less will be needed prior to the methanol synthesis. However, [14] concludes that this effect is comparatively small and atmospheric gasifiers have higher yields, and in total these systems result in higher efficiencies. In Ref. [14] it is concluded that applying the physical Selexol process for CO_2 removal increases the performance of the system compared to removal by MEA, since the heat demand of the MEA process strongly influences the possibilities for electricity production.

Not all studies report heat available for district heating (as shown by “?” in Fig. 3) but, as illustrated in our case study below, there is a trade-off between electricity production and excess heat available for DH or steam production. Most scenarios are net electricity consumers, although systems with net electricity production in combination with rather high methanol yield exist (nos. 10 and 18).

5.3. Case study – stand-alone gasification and MeOH production

The overall aim of our larger study, of which this paper is one part, is to evaluate commercial-scale gasification systems in terms of energy efficiency, climate impact and economy by case studies. The case studies will be made for different localisations and the relation to incumbent industries and infrastructure will be taken into consideration. A first step in making such analyses is to identify appropriate base configurations for the gasification system.

The aim of the case study in this paper is to show the impacts of process integrations and system boundaries for a stand-alone biomass gasification unit with methanol synthesis. There is also a setup where the methanol is used in a methanol-to-olefins (MTO) unit.

5.3.1. Process configuration – base case

Based on the analysis of previous studies and available data, the setup used by Isaksson et al. [11] was chosen for the case study. The performance in terms of energy efficiency and the technical maturity of the system configuration seem reasonable, given the size (430 MWth) and implementation time frame (2015–2020). The configuration is shown in Fig. 4.

In the case study system, the biomass is dried to 15% wt moisture content before entering the gasifier, which is a pressurised oxygen/steam-blown circulating fluidised-bed gasifier operating at 25 bar and 850 °C (based on Ref. [6]). The chosen gasification technology can be built in the desired size (430 MWth LHV biomass input at 50% moisture) and the injection of biomass to the gasifier will be possible without energy-demanding pre-treatment (like pyrolysis or torrefaction). The oxygen is provided by an ASU which is standard equipment. The product gas exiting the gasifier first passes through filters in order to remove particles and alkali before entering the scrubber. This is standard equipment for gas cleaning. Previous studies have shown that hot gas cleaning could increase the efficiency, e.g. [14,26], but such equipment is not yet on the market and was therefore not included in this study. Methane (and other hydrocarbons) in the syngas are removed by the oxygen-blown ATR operating at 25 bar and 1000 °C. The water–gas-shift changes the H_2/CO ratio to desired values (~ 2).[1] The recycle in the case of the LPMeOH does not lead to the same losses as in the case of conventional methanol synthesis [26]. Another reasonable choice of gasification technology would have been an indirect gasifier, but pressurising and scaling this technology is more uncertain.

5.3.2. Process description of MTO process

In addition to utilisation as motor fuel, methanol is a base chemical which can be used as feedstock for production of chemicals and materials. One interesting application is the methanol-to-olefins (MTO) process which converts methanol-to-olefins, mainly ethylene and propylene (Fig. 5). Currently there are two demonstration plants for the MTO processes in Norway. According to Chen et al. [33], the MTO process will be economically attractive especially at high crude oil prices (>US$ 30–40 bbl.). The effect on the energy balance of adding an MTO unit directly after the methanol synthesis was investigated for the stand-alone case.

The MTO process operates at temperatures between 300 and 600 °C and at slightly elevated pressures (1–3 bar). The estimates of yield and energy demand in the MTO process used in this study are based on Joosten 1998 [34] and Nouri and Tillman 2005 [35]. Joosten estimates the that the commercially viable MTO process would require 0.3 MJ electricity, 1.1 MJ high pressure steam and 2.8 MJ of external fuels to produce 2.11 kg olefins (47% ethylene and 53% propylene) from 6.3 kg methanol. Nouri and Tillman use the same estimates of energy demand and yields as [34] as an estimate also for the use of crude methanol (with ~20% water content) in the MTO process.

5.3.3. Energy performance of stand-alone gasification and methanol synthesis systems

Whether external or process-integrated drying is applied has a significant impact on the energy balance and efficiency of the system. As illustrated by Fig. 6, more excess heat is available for electricity and district heating production (or other utilisation) on the site of gasification if the biomass has been dried elsewhere

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[1] Note that with steam addition to the LPMeOH process there is no need for a separate WGS reactor (Hamelinck and Faaij, [26]). Isaksson [11] used a separate reactor in the simulation, but a setup without a separate WGS and direct steam addition to the LPMeOH would result in the same energy balance.

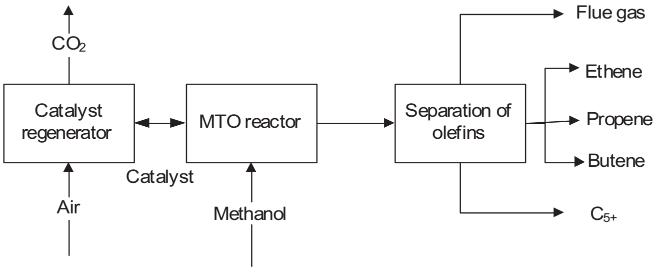

Fig. 5. Schematic description of MTO process. Source: [36].

(Fig. 6c and d), i.e. the cooling demand increases by 60% when excess heat from the process is not used for biomass drying. The efficiency of different drying technologies has been investigated by e.g. [37,38]. Heyne and Harvey 2009 [37] investigated three processes for biomass drying (air, steam and flue gas drying) and its process integration opportunities by pinch analysis to a biomass gasification process with downstream SNG-production. Their general conclusion is that process-integrated drying in the case of the SNG process can result in significant energy savings compared to a stand-alone case. Their specific conclusion is that steam drying has the highest potential for process integration with their SNG process. Fagernäs et al. [38] investigated biomass drying for second-generation synfuel production. The most promising commercially available drying processes were included in their study. They concluded that in large-scale steam cogeneration applications steam dryers may offer efficiency advantages, since it makes recovery of heat for DH possible.

Further Fig. 6 illustrates that heat pumping can improve the electricity production capacity of the system (diagram b). The effect of the heat pump is limited here but can be more important in other system configurations, e.g. as illustrated by Tock et al. [14] in a case with MEA for CO_2-separation.

By introducing more steam levels in the steam cycle, the electricity production and energy efficiency of the system can be increased; see Fig. 7. Tock et al. [14] show the benefits in electricity production of utilising expander, gas turbine (for off-gases) and advanced steam cycles, but conclude that the increase in overall efficiency is less than one per cent for a gasification system with DME synthesis when introducing a gas turbine compared to a case with only steam cycle. According to the same study the effect of introducing an advanced steam cycle is in the same range of efficiency improvement. In this study, off-gases are assumed to be burned in a boiler which enables heat-exchanging at high temperatures and thereby the high temperature of the superheated steam in the Rankine cycle. Heat-exchanging with the product gas at such temperatures is not possible today due to reducing conditions.

In Fig. 6, c and d show the trade-off in electricity and DH production depending on steam data for the steam cycle respectively. The steam data in c is the same as in a and b, whereas it has been adjusted in d to better fit the levels of available excess heat.

5.3.4. *Energy performance – including MTO production*

The limited information on the MTO process did not allow a full background–foreground analysis, but the steam and electricity demands of the process were taken into consideration (Fig. 8). The grand composite curve for the gasification and methanol process changed when the MTO process was added (Fig. 7a) since the distillation step could be removed.

5.3.5. *Utilisation of excess H_2 to increase methanol and process yield*

The grand composite curve (GCC) for the gasification and methanol production system with H_2 addition replacing the WGS is shown in Fig. 7b. The yield of methanol increases significantly, by

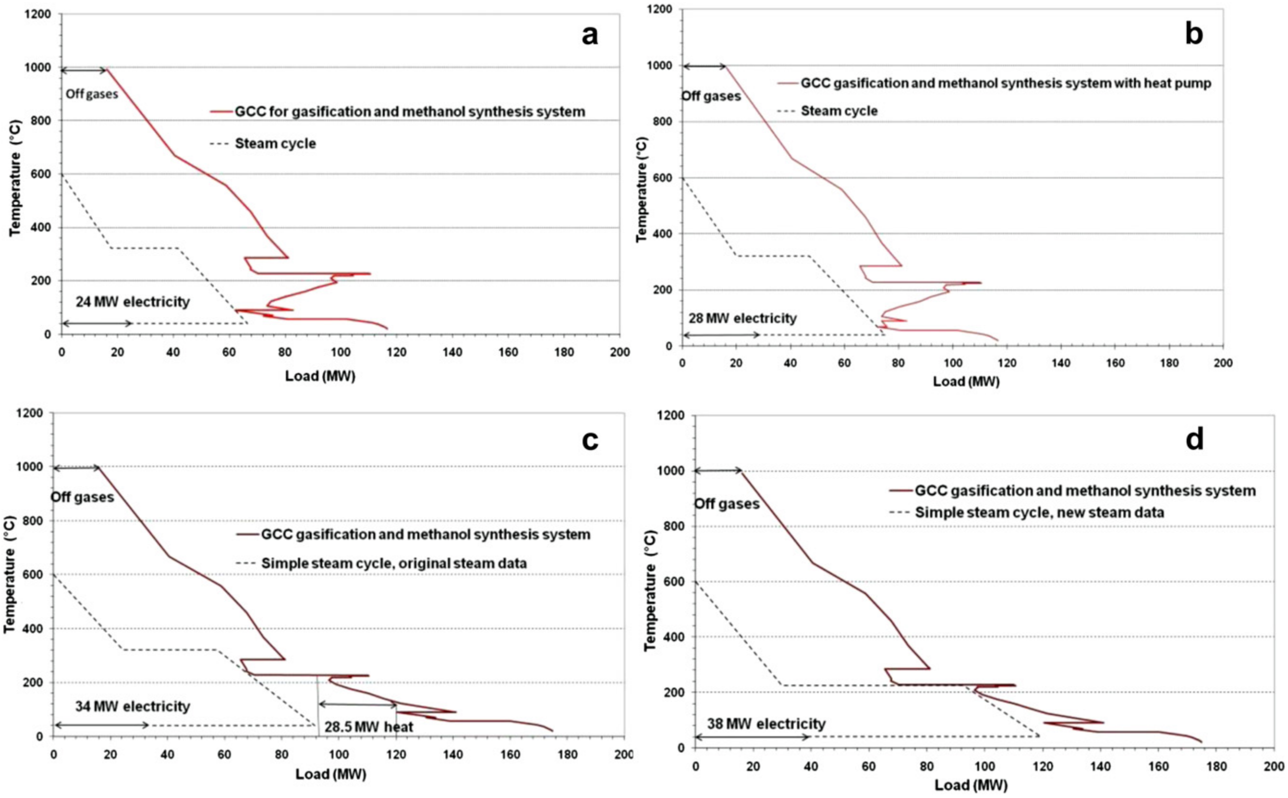

Fig. 6. Background–foreground analysis for steam cycle and GCC for stand-alone biomass gasification and methanol synthesis process. In a and b process-integrated biomass drying (flue gas drying) is included. In b a heat pump has been included. In c and d biomass drying is not included.

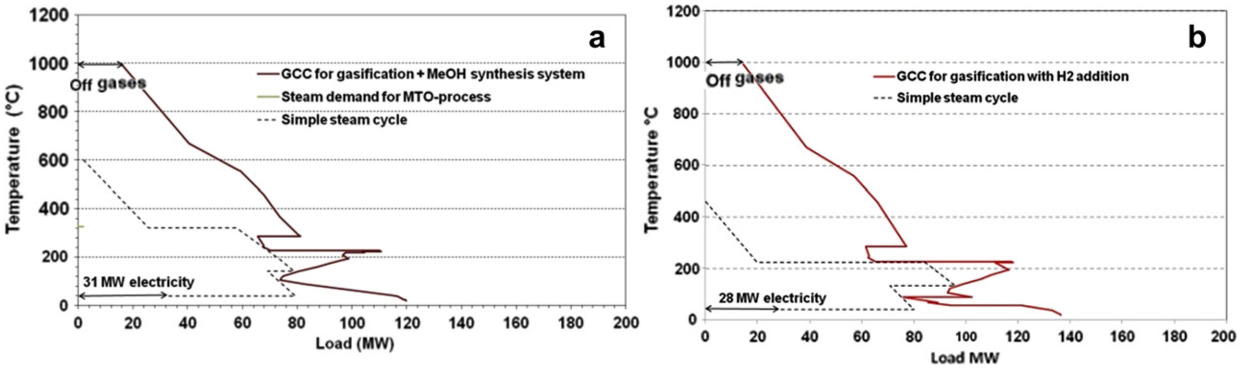

Fig. 7. Background–foreground analysis of steam cycle and GCC for stand-alone biomass gasification and methanol synthesis process. In a, an MTO process is also included, and in b, the WGS is replaced by H_2 addition.

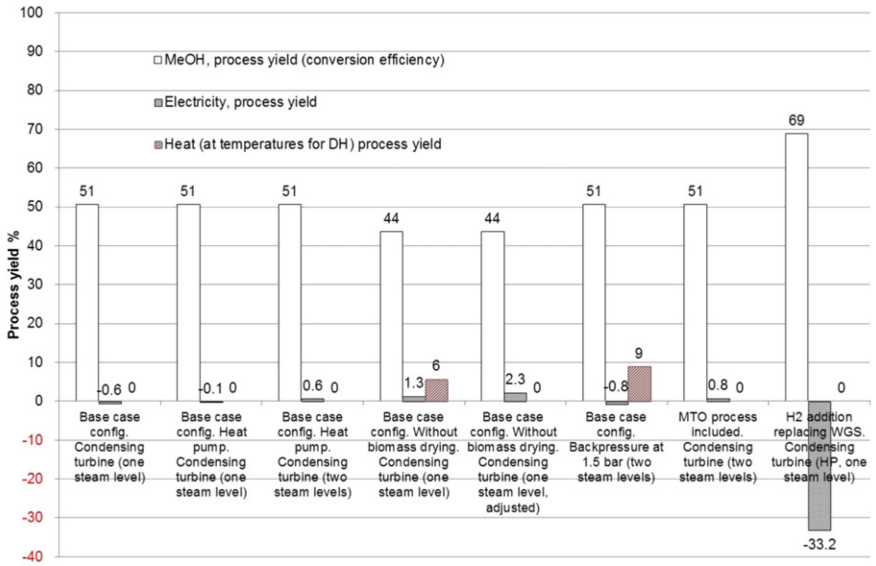

Fig. 8. Yield of methanol, electricity and heat (DH grade >100 °C) for the different configurations of the case study system. In all configurations, the starting point is wet biomass at 50% moisture content.

~35%, compared to having a shift reactor (Fig. 8). The drawback of such as system is the significant electricity demand for the production of the H_2. However, there might be sites with actual excess of H_2 which will not require additional electricity use for the production. The impact of locating the system at such a site could be interesting for future studies. In this study it was assumed that the H_2 was produced in an electrolyser with electricity demand according to [39]. The electrolyser could not fully cover for the oxygen demand, and an ASU producing some of the required oxygen was included.

5.3.6. *Results – product yields for different configurations of stand-alone case study*

According to Fig. 8 there is a clear trade-off in yields of methanol, electricity and heat, related to different choices in the configuration of the biomass gasification and methanol synthesis steps. The inclusion of different electricity production equipments (condensing, back-pressure, heat pumps etc.) is important for the outcome of the system.

As we concluded from the review of previous studies, the moisture content of the raw material has a significant impact on the calculated energy performance of the system. Therefore, the system boundaries in this study are set so as to include all drying of biomass starting with a moisture content of 50%. Hence, in cases where biomass drying occurs off-site, an estimated energy demand for this drying is still included in the system analysis. Fig. 8 shows that the cases with the lowest conversion efficiency from biomass to methanol are where biomass drying occurs off-site, requiring extra biomass.[2] These cases also have the highest net electricity production. The highest conversion efficiency from biomass to methanol is in the case where WGS is replaced by H_2 addition, but this case has a significant electricity deficit since the H_2 is assumed to be produced in an electrolyser. Note that in the case with MTO the methanol is an intermediate product and hence the final products are olefins.

6. Discussion

In this study and in most of the previous studies considering biomass gasification for methanol production, stand-alone configurations were considered. Biomass supply chains, prospects for district heating delivery and integration with incumbent industry are factors of importance that often are excluded. In the case study analysed in this paper, consistent system boundaries were applied and the energy demand for biomass drying was included also when not occurring on site. Drying the biomass elsewhere can reduce energy demand for transportation. However, the energy demand for on-site drying should be compared to the sum of the energy demand for off-site drying and energy savings from transporting dryer biomass. Depending on transportation modes, drying sources on- and off-site respectively, will determine on a case to case basis which choice that is optimal. Johansson et al. [40] estimate energy- and cost-savings for transportation due to drying of biomass to different moisture contents.

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[2] 3.1 MJ_{fuel}/kg H_2O evaporated, based on Heyne and Harvey 2009 [32].

In order to analyse optimal system solutions, economic and environmental evaluation should be added to the energy efficiency evaluation. Previous studies have shown that the evaluation of the energy efficiency can give one resultant optimal system whereas the economic evaluation can give another. Many studies use energy efficiency for multi-product systems (producing biofuels, electricity and heat) but the relevance and meaning of comparing energy of different qualities in one measure is questionable. The environmental and economic performances of the systems are important complements for determining optimal solutions.

7. Conclusions

From the reviewed studies, and from the case study, we see that there is a trade-off in yields of methanol, electricity and heat and thereby in energy balance for systems with different configurations of the biomass gasification and methanol synthesis steps.

From the analysis of the stand-alone case study, we conclude that whether or not biomass drying is considered has a major impact on the energy balance. In our case study the cooling demand increases by 60% when excess heat from the process is not used for biomass drying. If dry biomass is imported to the site, the excess heat from the process increases significantly; this increases the on-site electricity and DH production potential substantially. From previous studies and the case study, we conclude that process-integrated drying has a potential to increase the efficiency of the system.

H_2 addition replacing the WGS has a potential of increasing the methanol yield significantly (in the case study by ~35%), but in a stand-alone case the electricity demand for producing the hydrogen makes such a system unrealistic.

Adding an MTO process to the stand-alone methanol production showed little impact on the overall energy balance on the system. Steam and electricity are needed for the MTO process, but less is required in the methanol production since distillation can be omitted.

8. Future work

This study is part of a larger project analysing advantages of industrial cluster formation including commercial-scale biomass gasification systems producing second-generation biofuels or materials. The importance of process integration and incumbent infrastructure for the realisation of efficient systems (in terms of energy, greenhouse gas performance and economy), is investigated. The larger project includes several steps: energy analysis, environmental and economic evaluation. The results in presented in this paper will be used for making a comparison of energy efficiency, greenhouse gas emissions and economic performance for stand-alone and integrated biomass gasification methanol synthesis plants.

Acknowledgements

The authors would like to thank Johan Isaksson at the Division of Heat and Power Technology at Chalmers for prompt support on data retrieval. Financial support from the Swedish Energy Agency, Göteborg Energis Forskningsstiftelse, Preem, Perstorp Oxo, E.ON. Gasification Development Ltd, the Swedish Environmental Protection Agency and Formas is greatly acknowledged.

Appendix. Process configurations for biomass gasification systems with methanol synthesis, with scenario no. referring to Fig. 3.

Reference/ conditions	No.	Biomass input	Process configuration
Hamelinck and Faaij 2002 [26] Drying (30–15%) is considered	1	380 MW	Atm. indirect gasifier, scrubber, SMR, no shift, LPMeOH with steam addition & recycle, ST. No CO_2 removal prior to synthesis
	10	380 MW	Pressurised direct oxygen/ steam-blown gasifier, HGC, ATR, partial shift, conv. MeOH (with recycle), ST. Selexol CO_2 partially removed before synthesis
	7	380 MW	Atm. indirect gasifier, scrubber, SMR, partial shift, conv. MeOH & recycle. Selexol CO_2 removal. ST.
	2	380 MW	Pressurised direct oxygen/ steam-blown gasifier, HGC, ATR, no shift, LPMeOH with steam addition (no recycle), CC. No CO_2 removal prior to synthesis
	3	380 MW	Pressurised direct oxygen/steam-blown gasifier, scrubber, no reforming, no shift, LPMeOH with steam addition (no recycle), CC. No CO_2 removal prior to synthesis
Ecotraffic 1997 [41]	11	408 MW	Oxygen/steam-blown pressurised CFB gasifier. Recycle, shift, ATR, conv. MeOH. CO_2 removal prior to synthesis. Drying included. ST
	12	498 MW	Oxygen/steam-blown pressurised CFB gasifier. Recycle, shift, ATR, conv. MeOH. Integration with advanced heat and power plant $\eta el = 0.4$. CO_2 removal prior to synthesis. Drying included (from 50% moisture). GT + ST.
Brandberg et al. 2000 [29]	5	256 MW	Pressurised oxygen-blown CFB gasifier 20 bar 950 °C. Once through, GT + ST. LPMeOH. Selexol. Fluidised-bed dryer with MP steam as heat source, drying from 50% moisture content. ST in back-pressure mode
	6	256 MW	20 bar oxygen-blown CFB gasifier 950 °C. Once through, GT + ST. LPMeOH. Selexol. Fluidized bed dryer with MP steam as heat source; drying from 50% moisture content. ST in condensing mode
Tock et al. 2010 [14] Air drying (from 50% wt) + torrefaction included	8	20/400 MW	FICFB gasifier atm. WGS. ASU. Cold gas cleaning. Conv. MeOH synthesis. 90% gas recycle. MEA. ATR 950 °C, atm. GT + ST.
	9	20/400 MW	FICFB gasifier atm. WGS. ASU. Cold gas cleaning. Conv. MeOH synthesis. 90% gas recycle. Selexol. ATR 950 °C, atm. GT + ST
	13	20/400 MW	CFB gasifier 30 bar. WGS. ASU. Cold gas cleaning. Conv. MeOH synthesis. 90% gas recycle. MEA. ATR 1350 °C, 30 bar. GT + ST
	14	20/400 MW	CFB gasifier 30 bar. WGS. ASU. Cold gas cleaning. Conv. MeOH synthesis. 90% gas recycle. Selexol. ATR 1350 °C, 30 bar. GT + ST

(*continued*)

Reference/ conditions	No.	Biomass input	Process configuration
McKeough 2007 [32]	15	200–300 MW	Steam oxygen-blown pressurized FB gasification. Novel catalytic reforming, high temp. shift (performed before scrubbing), scrubber, regenerative CO_2 absorber. Long recycle.[a] Drying (30–15%) low pressure steam for drying. Conventional MeOH process
Isaksson et al. 2011 [11]	4	816 MW	Drying (50–15%) is included. Oxygen-blown pressurised CFB gasification. ASU. LPMeOH. 90 bar 240 °C. No CO_2 removal. Back-pressure in steam cycle due to steam demand for background process.
Clausen et al. 2010 [30] Drying 50–5% included.	22	17 MW	Atm. directly heated two-stage gasifier, scrubber. Electrolyser. Conv. MeOH synthesis 95% gas recycle. Steam drying.
	23	214 MW	Atm. directly heated two-stage gasifier, scrubber. Electrolyser. Conv. MeOH synthesis 95% gas recycle. MEA. Steam drying.
Sadhukhan and Ng 2011 [20] Bio-oil is raw material	19	675 MW	Pressurised EF gasifier, conv. MeOH. Once through. WGS. Electrolyser config. H_2 from electrolyser added to syngas. GT + ST
	20	1350 MW	Pressurised EF gasifier, conv. MeOH. Once through. WGS. Electrolyser config. H_2 from electrolyser added to syngas. GT + ST
	21	1350 MW	Pressurised EF gasifier, conv. MeOH. WGS. Recycle. Electrolyser config. H_2 from electrolyser added to syngas. GT + ST
	16	675	Pressurised EF gasifier, conv. MeOH. Once through. ASU config. WGS. GT + ST.
	17	1350	Pressurised EF gasification, conv. MeOH. Once through. ASU config. GT + ST.
	18	1350	Pressurised EF gasifier, conv. MeOH. Recycle. ASU config. GT + ST.

[a] In this recycle not only recycle in methanol synthesis is applied but also after shift to gasifier.

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